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COMPOSITIONS, ORIGINS, EMISSION RATES AND ATMOSPHERIC IMPACTS OF VOLCANIC GASES

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INTRODUCTION

Since the 18th Century, when Benjamin Franklin (1784) suggested that the 1783 Laki eruption in Iceland triggered an abnormally cold winter in 1783-4, many authors have noted a link between volcanic eruptions and short periods of cooler climate. At first, some linked the cooling to the large quantities of ash or dust injected into the atmosphere (Lamb, 1970). Now we know that injection of large quantities of volcanic sulfur gases (SO_2 and H_2S) into the stratosphere and the subsequent production of sulfuric acid aerosols generate volcanic climate forcing. This paper reviews recent research on the compositions, origins, emission rates, and atmospheric impacts of volcanic gases, and suggests possible avenues for future work.

COMPOSITIONS AND ORIGINS OF VOLCANIC GASES

Passively degassing and erupting volcanoes discharge magmatic gases and steam-rich gases from boiling meteoric and hydrothermal fluids into the atmosphere. Of these potential sources, degassing magma furnishes most of the volcanically derived pollutants (e.g., CO_2, SO_2, HCl, and HF). Under rare circumstances, these magmatic gases can be sampled directly from >500°C volcanic vents or fumaroles and brought back to the laboratory for analysis (Symonds et al., 1994). Results show that these high-temperature volcanic gases are dominated by H_2O, CO_2

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and SO_2 with lesser amounts of H_2, H_2S, HCl, HF, CO, S_2, COS, N_2, rare gases and a number of trace-metal species (e.g., CuCl, NaCl, H_2MoO_4, PbS, Hg) (Giggenbach and Matsuo, 1991; Symonds et al., 1994; Symonds and Reed, 1993). Several examples of high-temperature volcanic gases from terrestrial volcanoes are given in Table 1. These are some of the best available data because they represent equilibrium compositions at the reported temperatures. A more complete compilation of high-temperature volcanic gases in equilibrium is summarized by Symonds et al. (1994). The wide variation in the compositions of volcanic gases reflects variations in the tectonic setting, magma composition, degassing state of the magma, and the temperature and pressure of equilibration.

Table 1. Selected compositions of high-temperature volcanic gases from terrestrial volcanoes. Concentrations of species reported in mole%.

Volcano (Ref.)	T (°C)	H_2O	H_2	CO_2	CO	SO_2	H_2S	S_2	HCl	HF	COS
Mt. St. Augustine[1]	743	96.83	0.54	1.49	0.0060	0.22	0.38	---	0.51	0.025	---
Erta'Ale[2]	1130	77.24	1.39	11.26	0.44	8.34	0.68	0.21	0.42	---	0.0016
Mt. Etna[3]	1075	47.26	0.51	26.06	0.54	25.18	0.20	0.21	---	---	---
Mt. St. Helens[4]	710	98.6	0.39	0.886	0.0023	0.067	0.099	0.0002	0.076	0.03	1.8E-05
Kilauea[5]	1170	37.09	0.49	48.90	1.51	11.84	0.04	0.02	0.08	---	---
Kilauea[6]	997	81.6	0.9929	3.80	0.0702	12.0	0.761	0.358	0.171	0.20	0.0016
G. Merapi[7]	915	88.87	1.54	7.07	0.16	1.15	1.12	0.08	0.59	0.04	---
Nyiragongo[8]	970	43.50	1.29	48.55	2.20	2.02	1.72	0.62	---	0.09	0.0016
Poàs[9]	989	96.29	0.5240	0.7768	0.0066	1.511	0.0131	---	0.784	0.091	7.1E-08
Showa-Shinzan[10]	1015	98.04	0.63	1.2	0.0129	0.043	0.0004	2.6E-07	0.053	0.024	---
Surtsey[11]	1125	81.13	2.80	9.29	0.69	4.12	0.89	0.25	---	---	0.0016
Usu[12]	659	95.8	0.273	3.024	0.00440	0.258	0.609	0.0052	0.0241	0.0116	0.00032

--- not determined or below detection; [1]Sample Spine-1A (collected 7/6/89) from Symonds, Gerlach and Iven (unpublished); [2]Sample 910 (collected 1/23/74) from Giggenbach and Le Guern (1976) and Gerlach (1980b); [3]Sample #10 from hornito 2 (collected 7/12/70) from Huntingdon (1973) and Gerlach (1979); [4]Sample CNR (collected 9/17/81) from Gerlach and Casadevall (1986); [5]Sample J8 (collected 3/25/18) from Shepherd (1921), Jagger (1940), and Gerlach (1980a); [6]Sample Pele 4 (collected 1/14/83) from Gerlach (1993); [7]Sample Mer 79-2 (collected in 1979) from Le Guern et al. (1982); [8]Sample 2 (collected in 1959) from Chaigneau et al. (1960) and Gerlach (1980d); [9]Sample P44 (collected 6/19/81) from Delorme (1983) and Rowe (1991); [10]Sample from the A-1 vent (collected 9/8/54) from Nemoto et al. (1957) and Symonds et al. (1996); [11]Sample 12 (collected 10/15/64) from Sigvaldason and Elisson (1968) and Gerlach (1980c); [12]Sample 11 (collected 9/1/79) from Matsuo et al. (1982) and Gerlach (unpublished).

Subaerial explosive eruptions inject these magmatic gases directly into the atmosphere. However, at passively degassing subaerial volcanoes, magmatic gases sometimes interact with meteoric or hydrothermal water within the edifice of the volcano (Figure 1). Discharged gases

from submarine volcanoes also interact with seawater. These magmatic gas-water interactions trigger a series of scrubbing and precipitation reactions that may mask degassing of some or all species depending on (1) their water solubilities—SO_2, HCl, and HF are more soluble in water than CO_2 and H_2S— and (2) the gas-water ratio (Symonds and Gerlach, 1998).

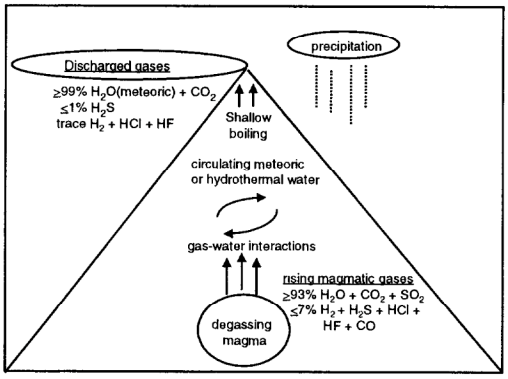

Figure 1. Schematic diagram showing how interactions between magmatic gases and meteoric or hydrothermal water within the edifice of a degassing volcano can alter the compositions of volcanic discharges.

Magmatic gas-water interactions greatly reduce the atmospheric impact of worldwide volcanic emissions, especially those from submarine volcanism. The gases discharging from mid-ocean ridges and other submarine volcanic activity, which currently supplies 80% of the magma to the earth's crust (Crisp, 1984), are mostly dissolved in the oceans. For example, although submarine volcanism emits 31-66 Mt y^{-1} of CO_2 or about half of the global volcanic emissions (Gerlach, 1991), virtually none of this CO_2 makes it to the atmosphere. Moreover, gas-water interactions clearly reduce or sometime eliminate magmatic gas emissions (e.g., especially SO_2, HCl, and HF) from many passively degassing subaerial volcanoes in repose between eruptions, although quantification of this process requires further study.

VOLCANIC EMISSIONS

To understand the impact of volcanic gases on the atmosphere, we need estimates of emission rates of the major species from present-day passively degassing and erupting volcanoes, and from large prehistorical eruptions that represent potential worst-case scenarios for future eruptions. Emissions of gases from presently degassing volcanoes can be measured remotely by airborne or ground-based methods, or by satellite. They can also be determined by direct sampling and profiling of volcanic plumes. Satellite methods work best for large explosive eruptions, whereas airborne and ground-based techniques work well for passively degassing volcanoes or for small eruptions where plumes fail to penetrate the tropopause. Due to available technology and gas-radiation absorption properties, the easiest volcanic-gas emission rate to measure is for SO_2; this can be determined remotely by airborne or ground-based techniques using the Correlation Spectrometer (COSPEC; Stoiber et al., 1983) and by satellite platforms equipped with the Total Ozone Mapping Spectrometer (TOMS; Krueger, 1983; Symonds et al., 1994). CO_2 emission rates can also be determined well using LI-COR technology, although this requires using a direct sampling and profiling approach that may limit its use under certain conditions (e.g., ash-laden plumes) (Gerlach et al., 1997). Fourier Transform Infrared Spectroscopy (FTIR) is an established technology just now showing promise in measuring emission rates of volcanic SO_2, HCl, and SiF_4 (Francis et al., 1998; McGee and Gerlach, 1998). Clearly, emissions of many volcanic-gas species cannot yet be measured directly. When emission rates of volcanic-gas species cannot be determined using direct instrumental techniques, they can be estimated using the emission rate of an easily determined species (e.g., SO_2) and compositional data on gas discharges (e.g., Table 1).

Unfortunately, determining gas emissions from eruptions before 1978 (e.g., pre-TOMS) is more difficult. Yet compared with eruptions this century, some prehistoric eruptions expelled colossal amounts of magma implying large stratospheric sulfur injections and huge climatic implications. The main methods to estimate volatile emissions from pre-1978 eruptions, both of which are often unsatisfactory, include determining the erupted volatile contents from petrologic studies of pre-erupted and erupted magma (the petrologic method; Devine et al., 1984) and measuring the volatile components in ice cores (Hammer et al., 1980). Although the petrologic method works well for basaltic eruptions where pre-eruption sulfur dissolves in the melt (Thordarson and Self, 1996), it underestimates the SO_2 released from many explosive eruptions involving evolved magmas (andesites, dacites, rhyolites) because these magmas are often gas-saturated prior to eruption (Gerlach et al., 1996; Scaillet et al., 1998). Ice-core estimates also suffer from serious problems including difficulties of determining the volcanic aerosol component in the core, high variability between cores even from the same area, dating uncertainties, and inconsistent snow deposition (Robock and Free, 1995). But recently some of these problems have been minimized by calibrating pre-1850 layers in ice cores with 1850-present optical depth data (Zielinski, 1995).

Table 2 gives examples of emission rates from passively degassing and erupting volcanoes, and estimates of the annual global volcanic emissions of CO_2, SO_2, HCl and HF. Although most of the emission rates represent only the past 20 years of degassing, the table also includes some data from prehistoric eruptions to demonstrate the magnitude of rare cataclysmic eruptions. Of course, SO_2 emission rates, especially from stratosphere-penetrating explosive volcanic eruptions (SPEVE), are most critical in determining the atmospheric impact of volcanic emissions. Although there are good TOMS measurements of SO_2 emissions for individual

SPEVE during the past 20 years (e.g., Table 2), it is much more difficult to quantify the annual SO_2 emission rate from SPEVE; some of the current estimates are 1.0 Mt (Pyle et al., 1996), 1.7 Mt (Stoiber et al., 1987), and 2.4 Mt (Bluth et al., 1993).

Table 2. Estimates of volcanic contributions of CO_2, SO_2, HCl, and HF to the atmosphere from individual volcanoes and from all present-day subaerial volcanoes.

Volcano and Reference	CO_2	SO_2	HCl	HF
Eruptive Degassing Emission Rates (Mt/eruptive event)				
Rosa flood basalt (14 Ma)[1]	---	12,420	710	1780
Toba (74 ka)[2]	---	6500	---	---
Pinatubo (6/15/91)[3]	42	17	3	---
El Chichón (4/4/82)[4]	---	7	---	---
Mount St. Helens (5/18/80)[4]	---	1.0	---	---
Passive Degassing Emission Rates (t/day)				
Mount Etna (1975-87)[5]	63000	4000	1300	160
Popocatépetl (1/5/95 - 1/10/95)[6]	9000	3100	---	---
Oldoinyo Lengai (6/94)[7]	6600	8.0	1.5	0.54
Mount St. Helens[8]	4800	400	---	---
White Island[9]	1800	620	190	5
Kilauea (2/13/84)[10]	1300	220	---	---
Global Volcanic Emission Rates (subaerial volcanism; Mt/year)				
Williams et al. (1992)	64	---	---	---
Gerlach (1991)	86	---	---	---
Graf et al. (1997)	---	28	---	---
Bluth et al. (1993)	---	13	---	---
Stoiber et al. (1987)	---	18.7	---	---
Berresheim and Jaeschke (1983)	---	15.2	---	---
Symonds et al. (1988)	---	---	0.4 - 11	0.06 - 6

--- not determined; [1]Data for the Rosa member of the Columbia River Basalt Group (Thordarson and Self, 1996); [2]SO_2 emissions estimated from the H_2SO_4 emission rate of Chesner et al. (1991); [3]Data from Gerlach et al. (1996), Bluth et al. (1992), Read et al. (1993), McPeters (1993); [4]SO_2 emission rate recalculated by Bluth et al. (1997) using version 6 of the TOMS algorithm.; [5]Average CO_2 and SO_2 emission rate for the 1975-87 period from Allard et al. (1991), assuming that 44% of the total CO_2 emissions are from diffusive degassing, and HCl and HF emission rates for July 1987 from Andres et al. (1993); [6]Best estimate of average SO_2 and CO_2 emission rates for January 5-10, 1998 from Gerlach et al. (1997); [7]Koepenick et al. (1995).; [8]Average CO_2 and SO_2 emission rates for July 1980 to September 1981 from McGee and Casadevall (1994); [9]Rose et al. (1986) [10]Greenland et al. (1985).

ATMOSPHERIC IMPACTS

Emissions from SPEVE produce greater atmospheric impacts than tropospheric emissions from smaller explosive eruptions and passively degassing volcanoes that are removed rapidly by precipitation. But some speculate that tropospheric emissions from voluminous flood basalt eruptions may also cause significant climatic effects due to the shear volume of gas emissions (Thordarson and Self, 1996). Figure 2 summarizes the atmospheric impacts of large explosive eruptions. These eruptions inject gases (e.g., table 1) and ash 15 to 40 km above the Earth's surface into the stratosphere. The most significant consequence of these stratospheric volcanic emissions is the conversions of SO_2 to sulfuric acid aerosols. The total volume of injected aerosols and their average size affect their ability to scatter, both elastically and inelastically, short-wavelength (visible) solar radiation and long-wavelength (infrared) terrestrial radiation and hence, their capacity to alter the radiation balance of the atmosphere (Lacis et al., 1992). Due to their small size, volcanic aerosols are better at backscattering incoming visible radiation (e.g., increasing albedo) than at absorbing outgoing infrared radiation which results in net tropospheric cooling (Minnis et al., 1993). For instance, the 15 June 1991 eruption of Mount Pinatubo resulted in global cooling of about 0.5°C for a few years after the eruption (Hansen et al., 1992).

In addition to producing tropospheric cooling, volcanic aerosols also warm the stratosphere by absorbing solar and terrestrial radiation (Newell, 1970; Angell, 1993). Moreover, in a manner similar to polar stratospheric clouds (Solomon, 1990), volcanic aerosols form active surfaces for complex heterogeneous reactions between various stratospheric nitrogen and chlorine species; these reactions promote the destruction of ozone by converting inert stratospheric Cl species, elevated by anthropogenic chlorofluorocarbons, into ozone-destroying ClO (Hofmann et al., 1994). With time, the sulfuric acid aerosols coagulate and descend to the

upper troposphere where they form cloud condensation nuclei for cirrus clouds; these may further alter the atmosphere's radiation balance, although determining their effect on climate requires more work (Sassen, 1992).

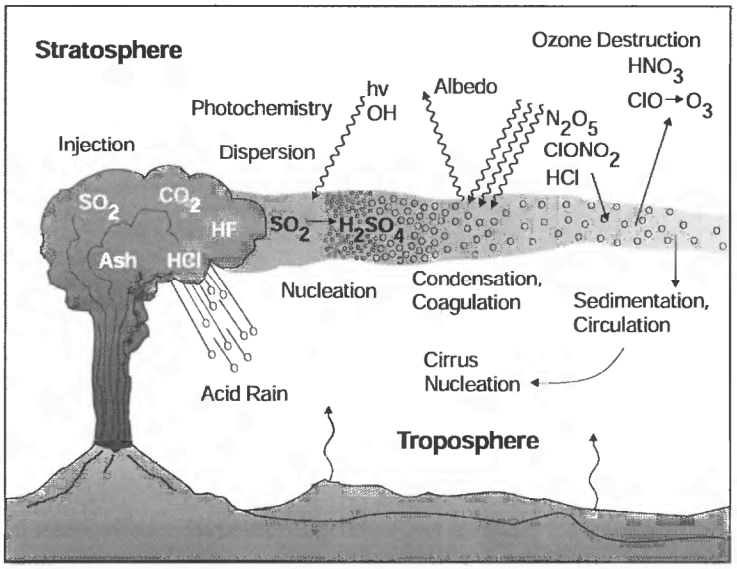

Figure 2. Schematic diagram showing the main atmospheric effects of large explosive eruptions that penetrate the stratosphere. After McGee et al. (1997).

Large explosive eruptions may also release significant amounts of the greenhouse gas, CO_2, although the average annual volcanic CO_2 emissions estimated at 86 MT y^{-1} are dwarfed by the anthropogenic CO_2 emissions of 26,000 MT y^{-1} (Gerlach, 1991). Stratospheric-penetrating eruptions also sometimes discharge significant amounts of HCl and HF (Table 2). Past studies (Johnston, 1980; Symonds et al., 1988) suggest that stratospheric HCl injection by large volcanic eruptions could enhance global ozone destruction, but recent work by Tabazadeh and Turco (1993) suggests that most of the HCl in eruption plumes is removed prior to stratospheric injection by dissolution in condensed supercooled water droplets followed by precipitation or scavenging by falling ash particles. HF might also be removed by similar processes.

DIRECTIONS FOR FUTURE WORK

Although the past twenty five years produced many advances in estimating volcanic gas contributions to the atmosphere, we still have several remaining hurdles. Future work on volcanic emissions should focus on three key areas: (1) Developing new technologies for remote measurements of CO_2, H_2S, HCl, and HF. Currently, CO_2 emission rates can be measured with LI-COR technology, which requires direct plume sampling and profiling. Although emissions of HCl and HF can sometimes be estimated by combining plume data on HCl/SO_2 and HF/SO_2 with COSPEC SO_2 emission rates, instrumental methods are unavailable for measuring H_2S and HF, and FTIR methods for HCl are still maturing. Remote measurements of CO_2, H_2S, HCl, and HF emissions are much preferred because direct sampling of plumes is not always possible, especially during eruptions when emission rates are at their highest levels. (2) Improving the volcanic emissions database; this includes measuring emissions for new species as well as from new volcanoes. Until recently, most volcanic emission-rate studies have focused on measuring SO_2 emissions. But for understanding the atmospheric impact of volcanic emissions, it is also important to obtain data for CO_2, H_2S, HCl, and HF. We also need to make new and existing technologies widely available to all countries that have degassing volcanoes. (3) Obtaining more reliable and complete data on emissions from large eruptions, especially prehistoric eruptions that are several orders of magnitude larger than historical events. Large eruptions, especially explosive events that penetrate the stratosphere, have the largest impact on climate. With the advent of TOMS, we have a good understanding of volcanic climate forcing by explosive eruptions in the past two decades. But we still need a better understanding of the climate implications for much larger prehistoric eruptions.

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